

Characterizing critical exponents via Purcell effect

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We investigate the role of phase transitions into the spontaneous emission rate of quantum emitters embedded in a critical medium. Using a Landau-Ginzburg approach, we find that, in the broken symmetry phase, the emission rate is reduced or even suppressed due to the photon mass generated by the Higgs mechanism. Moreover, we show that the spontaneous emission presents a remarkable dependence upon the critical exponents associated to a given phase transition, allowing for an optical determination of the universality class. Our findings not only demonstrate that the Purcell effect constitutes an efficient optical probe of distinct critical phenomena, but they also unveil that a more general connection between phase transitions and spontaneous emission exist, as previous experimental and numerical evidences suggest.

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Critical phenomena and phase transitions are amongst the most important and interdisciplinary research areas in physics. Criticality is known to dramatically affect many structural, thermal, and electrical properties of matter [1]. The importance of the concept of criticality extrapolates the domains of physics and finds applications in mathematics, biology, chemistry, and even economy and social sciences [1]. In addition to its phenomenological relevance, the field of critical phenomena has always been the scenario of new and groundbreaking theoretical ideas over the years, such as renormalization group and topological phase transitions [2].

In optics, critical phenomena in matter also show up in a crucial way. Important examples are the optical bistability [3], the many optical manifestations of structural phase transitions in liquid crystal [4] and, more recently, the optical analogue of the spin glass phase transition in random [5, 6] and homogeneous lasers [7]. Besides, the development of structured, artificial material platforms to investigate light-matter interaction, such as photonic crystals and metamaterials, has opened new venues to investigate optical manifestations of phase transitions. For instance, manifestations of the percolation phase transition were experimentally shown to occur in the Fano lineshape that describes light reflection upon disordered photonic crystals [8].

The high sensitivity of the spontaneous emission (SE) rate of an excited dipole emitter to the local environment makes the Purcell effect [9] especially prone to be influenced by phase transitions in matter. Indeed, the Purcell effect and single-molecule spectroscopy are unique tools to locally probe the electromagnetic environment at the nanoscale [10], with applications in solar cells [11], molecular imaging [10, 12], and single-photon sources [13]. In addition, progress in the field of nanophotonics and metamaterials has allowed for unprecedented control of the SE rate in artificial media such as invisibility cloaks [14], graphene-based structures [15], nanoantennas [16], photonic crystals [17], and hyperbolic metamaterials [18]. In particular, the latter may undergo a topological phase

transition that manifests itself in the Purcell factor [19]. By inducing long-range spatial correlations, structural phase transitions were demonstrated to have a dramatic impact on the distribution of decay rates in disordered photonic media [20]. The decay rate of emitters embedded in a medium undergoing a structural phase transition induced by the temperature is also characteristically affected at criticality, even though other optical phenomena such as light scattering are insensitive to phase-switching behaviour [21]. Another example is the percolation transition, which was shown to largely enhance the decay rate of quantum emitters and crucially govern the decay pathways [22]. In addition, fluctuations of the local density of states were experimentally shown to be maximum in thin metallic films near the percolation transition [23]. Altogether these recent findings on the Purcell effect at phase transitions, of different physical origins, suggest that a more general and profound connection between these phenomena exists.

In order to elucidate this issue, in the present Letter we investigate the effects of a phase transition into the SE rate of emitters when embedded in a bulk critical medium. By means of a generic Landau-Ginzburg description we find, without specifying any particular physical system, that in the broken symmetry phase, the emission rate is reduced (or even suppressed) due to the photon mass generated by the Higgs mechanism. Moreover, we show that the spontaneous emission presents a remarkable dependence upon the critical exponents associated to a given phase transition, allowing for the determination of the universality class in the broken symmetry phase. In the symmetric phase, we show that enhanced critical fluctuations lead to an anomalously large enhancement of the SE at critical point.

The Purcell effect is characterized by an enhancement of the spontaneous emission rate, Γ , of atoms or molecules by its environment. For a two-level system in free space, the SE rate is

$$\Gamma_0 = \frac{\omega_0^3 \mu^2}{3\pi\epsilon_0 \hbar c^3} = \frac{\pi\omega_0 \mu^2}{3\epsilon_0 \hbar} \rho_0(\omega_0) \quad (1)$$

where ω_0 is the two-level transition frequency, $\boldsymbol{\mu}$ is the transition dipole moment, and we identified the local density of states (LDOS) in vacuum $\rho_0(\omega) = \omega^2/\pi^2 c^3$. Changes in the electric dipole coupling and/or boundary conditions usually modify Eq. 1, which can be more easily identified by rewriting the SE rate (divided by 2π) as

$$g^2 \rho_0(\omega) \equiv \frac{1}{2\hbar\epsilon_0} \sum_{\mu,\nu=1,2} \int \frac{d^3\mathbf{k}}{(2\pi)^3} |\hat{\mathbf{e}}_{\mathbf{k}} \cdot \boldsymbol{\mu}|^2 \omega_{\mathbf{k}}^2 \mathcal{A}_{\mu\nu}^{(0)}(\omega_{\mathbf{k}}, \omega), \quad (2)$$

where g^2 is a convenient normalization factor, $\omega_{\mathbf{k}} = |\mathbf{k}|c$, $\hat{\mathbf{e}}_{\mathbf{k}}$ is the polarization versor, and $\mathcal{A}_{\mu\nu}^{(0)}(\omega_{\mathbf{k}}, \omega)$ is the free-photon spectral function

$$\begin{aligned} \mathcal{A}_{\mu\nu}^{(0)}(\omega_{\mathbf{k}}, \omega) &= -\frac{1}{\pi} \lim_{\delta \rightarrow 0} \text{Im} G_{\mu\nu}^{(0)}(\omega_{\mathbf{k}}, \omega + i\delta) \\ &= \frac{\delta_{\mu\nu}}{\omega_{\mathbf{k}}} \{ \delta(\omega - \omega_{\mathbf{k}}) - \delta(\omega + \omega_{\mathbf{k}}) \}, \end{aligned} \quad (3)$$

obtained from the free-photon propagator (we use the Feynman gauge)

$$G_{\mu\nu}^{(0)}(k^2) = \frac{i\eta_{\mu\nu}}{k^2}, \quad (4)$$

where $k = (k_0, \mathbf{k})$, $\eta_{\mu\nu} = \text{diag}(1, -1, -1, -1)$, and we used Lorentz invariance to simplify the dependence of $G_{\mu\nu}^{(0)}$ to k^2 . In fact, it is easily seen that substitution of (3) into (2) leads to (1).

Equation (2) relates the SE rate directly to a property of its environment, in this case, the Green function of the quantized electromagnetic field in free space. For interacting fields, a natural generalization for the electromagnetic contribution for the SE rate then is

$$\Gamma \equiv 2\pi g^2 \rho(\omega_0). \quad (5)$$

where $\rho(\omega)$ is now given by an analogue of Eq.(2), but with $\mathcal{A}_{\mu\nu}^{(0)}$ replaced by the interacting electromagnetic spectral function $\mathcal{A}_{\mu\nu}$.

With all that in mind, we now can describe our system. The emitter is embedded in a bulk critical medium so that the Lagrangian reads

$$\mathcal{L} = -\frac{1}{4} F_{\mu\nu} F^{\mu\nu} + \frac{1}{2} |D_\mu \varphi|^2 - a(T) \varphi^* \varphi - b(\varphi^* \varphi)^2. \quad (6)$$

Here φ is a complex-scalar order parameter that couples to the electromagnetic field through the covariant derivative $D_\mu = \partial_\mu - ieA_\mu$, and the field strength tensor is $F_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$. As usual, $a(T)$ is a function of $(T - T_c)$ and changes sign at the transition $T = T_c$; $a(T)$ and $b > 0$ are the parameters that are used to label the different phases of the system, where T is the temperature.

We proceed by: i) calculating $G_{\mu\nu}(k^2)$ taking into account the environment and/or boundary conditions; ii)

extracting, from it, $\mathcal{A}_{\mu\nu}(\omega_{\mathbf{k}}, \omega)$ and then $g^2 \rho(\omega)$; iii) obtaining Γ from Eq. (5).

When $T < T_c$ and $a(T) < 0$, the φ field acquires a nonzero vacuum expectation value, $\varphi_0^2 = -a/2b = v^2$, and we need to consider perturbations around the symmetry broken vacuum, $\varphi(x) = e^{i\frac{\theta(x)}{v}} [v + \rho(x)]$, where ρ and θ describe longitudinal and transverse fluctuations of the order parameter φ . In terms of these quantities

$$\begin{aligned} \mathcal{L} &= -\frac{1}{4} F_{\mu\nu} F^{\mu\nu} + \frac{M^2}{2} A_\mu^2 \\ &+ \frac{1}{2} (\partial_\mu \rho)^2 + \frac{m_\rho^2}{2} \rho^2 + \frac{1}{2} (\partial_\mu \theta)^2 + \dots \end{aligned} \quad (7)$$

with the mass $m_\rho^2 = 2|a(T)|$ being the mass of the longitudinal mode, ρ , while the transverse fluctuations, θ , are massless, in accordance to Goldstone's theorem [24]. Note that a nonzero expectation value $v \neq 0$ provides the gauge field, A_μ , with a mass, $M = ve$. This is the so called Higgs mechanism [24], in which case the massive photon propagator becomes

$$G_{\mu\nu}^M(k^2) = \frac{i\eta_{\mu\nu}}{k^2 - M^2 c^4 / \hbar^2}, \quad (8)$$

so that the photon spectral function reads

$$\mathcal{A}_{\mu\nu}^M(\omega_{\mathbf{k},M}; \omega) = \frac{\delta_{\mu\nu}}{\omega_{\mathbf{k},M}} \{ \delta(\omega - \omega_{\mathbf{k},M}) - \delta(\omega + \omega_{\mathbf{k},M}) \}, \quad (9)$$

where the dispersion relation is $\omega_{\mathbf{k},M} = \sqrt{c^2 |\mathbf{k}|^2 + M^2 c^4 / \hbar^2}$. The appearance of a mass term for the photons not only shifts the position of the poles in the spectral function, but, more importantly, reduces its spectral weight. After calculating $g^2 \rho(\omega)$ from Eq. (2) and using Eq. (5) with $\mathcal{A}_{\mu\nu}^M$ given by Eq. (9), we obtain

$$\Gamma = \frac{\omega_0^3 \mu^2}{3\pi \epsilon_0 \hbar c^3} \sqrt{1 - \frac{M^2 c^4}{\hbar^2 \omega_0^2}} = \Gamma_0 \sqrt{1 - \left(\frac{Mc^2}{\hbar \omega_0} \right)^2}. \quad (10)$$

A nonzero photon mass reduces the value of the SE rate in the Higgs phase, and even suppresses it, for $\hbar \omega_0 < Mc^2$, when the energy $\hbar \omega_0$ is not large enough as to overcome the rest energy Mc^2 , see Fig. 1. It is important to remark that this result is valid regardless the specific form of the parameter $a(T)$ as long as it changes sign at T_c . Assuming a typical power law dependence $M(t) = M_0 |t|^\beta$ (with $t = 1 - T/T_c$), our findings show that, in the broken Higgs phase ($T < T_c$), the SE rate increases with T and its behaviour crucially depends on the value of β (see Fig. 2). As a result, critical exponents β may be easily distinguished as their effects on SE rate are present not only close to the transition ($T \sim T_c$), but throughout $0 < T < T_c$. Altogether, our results demonstrate that one can determine the universality class of an arbitrary phase transition. This can be seen in Fig. 2

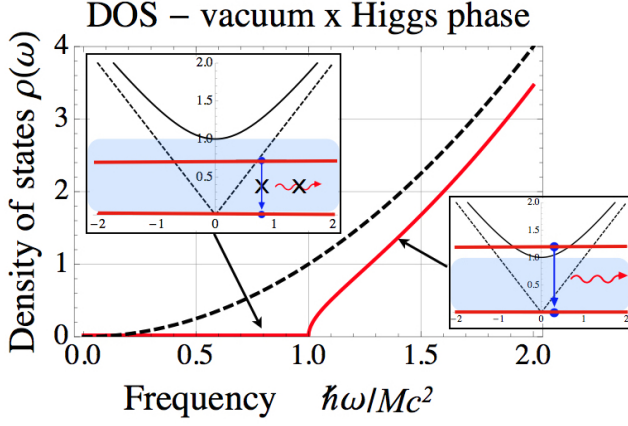


FIG. 1: (Main figure) photon DOS for the vacuum (black dashed curve) and Higgs phases (solid red curve). (Insets) for $\hbar\omega_0 < Mc^2$, inside the shaded area, no photons are available and no emission occurs; for $\hbar\omega_0 > Mc^2$, outside the shaded area, the DOS is finite (however low) and emission is allowed.

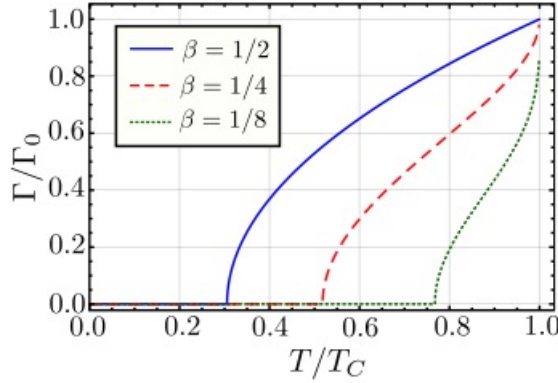


FIG. 2: The ratio Γ/Γ_0 at the Higgs phase ($T < T_c$) for different typical values of the critical exponent $\beta = 1/2, 1/4$, and $1/8$.

where the SE rate is calculated for typical values of β , such as $\beta = 1/2$ (mean field), $\beta = 1/8$ (Ising model), $\beta = 1/4$.

We now focus on the behaviour of the SE rate in the symmetric phase. For $T > T_c$ and $a(T) > 0$, there is no spontaneous breakdown of the vacuum symmetry so $v = 0$ and then

$$\mathcal{L} = -\frac{1}{4}F_{\mu\nu}F^{\mu\nu} + \frac{1}{2}|D_\mu\varphi|^2 - \frac{m^2}{2}\varphi^*\varphi - b(\varphi^*\varphi)^2, \quad (11)$$

where $m = 2a(T) > 0$, for all components of φ , and we see that the photon is massless. Thus, in this phase, the presence of a medium surrounding the emitter does not lead to a position shift of the pole in the photon propagator, but it rather renormalizes the vacuum polarization. There are two interaction vertices that contribute to the vacuum polarisation [25]

$$-ie(\varphi^*\partial_\mu\varphi A_\mu - (\partial_\mu\varphi)^*\varphi A_\mu), \quad \text{and} \quad 2ie^2\varphi^*\varphi A_\mu^2. \quad (12)$$

Lorentz invariance, however, constrains the vacuum polarization to have a precise form [24]

$$i\Pi_{\mu\nu}(q^2) = iq^2\eta_{\mu\nu}\Pi(q^2), \quad (13)$$

where the second order contribution is shown in Fig. 3.

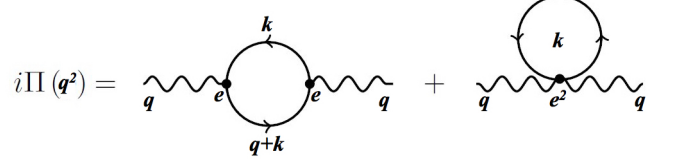


FIG. 3: Vacuum polarization, $\Pi(q^2)$, up to order e^2 , from fluctuations of the complex scalar order parameter, φ^*, φ .

Going beyond the second order by summing the non-1PI iterations of the graphs in Fig. 3, leads to the renormalized photon propagator

$$\begin{aligned} G_{\mu\nu}^Z(q^2) &= G_{\mu\nu}^{(0)}(q^2) + G_{\mu\alpha}^{(0)}(q^2)\Pi_{\alpha\beta}(q^2)G_{\beta\nu}^{(0)}(q^2) + \dots \\ &= Z(q^2)G_{\mu\nu}^{(0)}(q^2), \end{aligned} \quad (14)$$

with

$$Z(q^2) = \frac{1}{1 - \Pi(q^2)}. \quad (15)$$

The function $\Pi(q^2)$ is well known and it is regular at $q = 0$, thus ensuring that the photon remains massless. In the limit $q^2 \gg m^2$, and setting $q^2 = \Lambda^2$, it is given by [24]

$$\Pi(q^2 = \Lambda^2) = \frac{\alpha}{12\pi} \ln(\Lambda^2/m^2), \quad (16)$$

where Λ is a characteristic momentum scale in the problem and $\alpha = 1/137$ is the fine structure constant.

For the case of standard quantum electrodynamics, where $m = m_e$ is the electron mass, one usually choses the *on shell* renormalization point, $\Lambda = m_e$, in such a way that $\Pi(q^2 = m_e^2) = 0$ and $Z(q^2 = m_e^2) = 1$. In solid state systems, however, where the mass term corresponds instead to the inverse of a correlation length, $m = \xi^{-1}$, the scale Λ corresponds to a given natural cutoff in the problem (e.g., the Fermi momentum k_F , or the Debye momentum q_D , or, yet, the inverse lattice spacing $1/a$, among others). Here we choose $\Lambda = 1/\xi_0$, where ξ_0 is some fixed length scale far away from the critical point, $\xi(T \gg T_c) \approx \xi_0$, in such a way that, for $T \gg T_c$, we end up with $\Pi(q^2 = \xi_0^{-2}) = 0$ and $Z(q^2 = \xi_0^{-2}) = 1$.

The photon spectral function can be calculated as

$$\begin{aligned} \mathcal{A}_{\mu\nu}^Z(\omega_{\mathbf{k}}; \omega) &= -\frac{1}{\pi} \lim_{\delta \rightarrow 0} \text{Im} G_{\mu\nu}^Z(\omega_{\mathbf{k}}, \omega + i\delta) \\ &= Z(\xi^2) \mathcal{A}_{\mu\nu}^{(0)}(\omega_{\mathbf{k}}; \omega), \end{aligned} \quad (17)$$

and, as a consequence, the SE rate becomes

$$\Gamma = Z(\xi^2/\xi_0^2) \frac{\pi\omega_0\mu^2}{3\varepsilon_0\hbar} \rho(\omega_0) = Z(\xi^2/\xi_0^2)\Gamma_0. \quad (18)$$

We clearly see that for $T \gg T_c$, when $\xi \rightarrow \xi_0$ and $Z \rightarrow 1$, we have $\Gamma \rightarrow \Gamma_0$. As the critical point is approached, where $T \rightarrow T_c$ and $\xi \gg \xi_0$, one finds $Z \gg 1$, leading to a large enhancement of the SE rate. The divergence in the SE rate, Eq. (18), can be understood as a result of the unscreening of the electric charge when $\xi \rightarrow \infty$.

The correlation length ξ may approach criticality through a power law $\xi(t) = A|t|^{-\nu} + \xi_0$, or through another function such as $\xi(t) = \xi_0 \exp(\delta/|t|)$. Due to the logarithmic dependence in Z , any choice of power law for ξ in the symmetric phase does not lead to significant effects. In order to illustrate that, in Fig. 4 we plot the normalized SE rate with $M(t) = M_0|t|^{1/2}$ in the Higgs phase, and $\xi(t) = \xi_0 \exp(10/|t|)$ for the symmetric phase (recalling that $t = 1 - T/T_c$). It is clear that, coming from the symmetric phase, the enhancement is huge close to T_c .

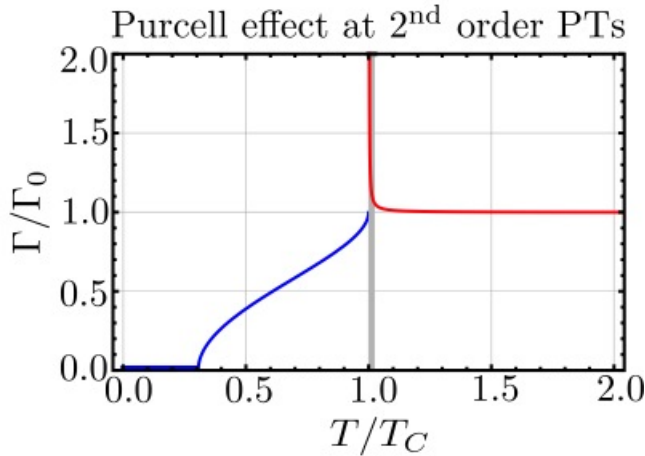


FIG. 4: Purcell effect close to a 2nd order PT at T_c . The ratio Γ/Γ_0 is shown for the two phases: i) Higgs phase (in blue) both for $\hbar\omega_0 < M(T)c^2$, where $\Gamma = 0$, or $\hbar\omega_0 > M(T)c^2$, where $\Gamma < \Gamma_0$; ii) symmetric phase (in red), where Γ diverges as $\xi \rightarrow \infty$, close to the stable IR fixed point.

In conclusion, we have investigated, using a Landau-Ginzburg approach, the effects of phase transitions in the SE rate of quantum emitters embedded in a critical medium. In the broken symmetry phase, we demonstrate that the SE rate is reduced and even suppressed due to the photon mass generated by the Higgs mechanism. In this same phase we show that the SE crucially depends on critical exponents associated to a given phase transition, without specifying *a priori* any particular physical system. This fact allows one to determine the universality class of phase transitions by optical means. Our results show that a general connection between critical phenom-

ena and the Purcell effect exists, as previous experimental and numerical evidences, involving different phase transitions, suggest [20–23]. Hence our analysis provide a qualitative theoretical basis for these findings, revealing they are actually deeply related. Altogether, our work not only unveils the general connection between critical phenomena and the Purcell effect, but it also demonstrates that the latter may be exploited as an optical probe of phase transitions and their universality classes.

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